

Accumulation and extraction of ultracold neutrons from a superfluid helium converter coated with fluorinated grease

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February 4, 2008

Abstract

We report experiments on the production of ultracold neutrons (UCN) in a converter of superfluid helium coated with fluorinated grease. We employed our technique of window-free extraction of accumulated UCN from the helium, in which they were produced by downscattering neutrons of a cold beam from the Munich research reactor. The time constant for UCN passage through the same extraction aperture as in a previous experiment was a factor two shorter, despite a lower mean velocity of the accumulated UCN in the present experiments. A time-of-flight measurement of the cold neutron spectrum incident on the converter allowed us to estimate the multi-phonon contribution to the UCN production. The UCN production rate inferred from two methods agrees with the theoretical expectation.

PACS numbers: 78.70.Nx, 28.20.Fc, 29.25.Dz, 61.12.Ha

Keywords: ultracold neutrons, UCN, UCN sources

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1 Introduction

Ultracold neutrons (UCN) have energies in the neV range and velocities up to a few meters per second. When impinging on suitable materials they undergo total reflection under any angle of incidence and can therefore be trapped in bottles and manipulated for a long time (see the books [1, 2] for an introduction to the physics of UCN). Owing to this feature UCN have become very useful in various fundamental investigations of neutron properties, with strong implications for particle physics and cosmology. The longstanding search for the neutron electric dipole moment investigates CP-violation beyond the standard model of particle physics [3, 4]. The accurate determination of the neutron lifetime is required for a detailed understanding of big bang nucleosynthesis [5], and to investigate strength and structure of the semi-leptonic weak interaction within the first quark family (see, e.g., the workshop proceedings [6, 7]). New topics with UCN are, among others, the demonstration of quantum levels of neutrons in the earth's gravitational field [8], and even more recently, the search for neutron - mirror neutron transitions [9, 10]. The most intense UCN source at the Institut Laue Langevin in Grenoble [11] provides

not more than 50 UCN per cm^3 . In order to improve counting statistics, new UCN sources are being developed in many laboratories around the world [12, 13, 14, 15, 16, 17, 18].

An elegant method to produce UCN employs a converter of superfluid ^4He in a beam of cold neutrons [19]. The kinematics defined by the dispersion relations of helium and the free neutron enables downscattering of cold neutrons with an energy around 1.0 meV (wavelength 0.89 nm) to ultracold energies via emission of a single phonon. In addition, multi-phonon processes occur which contribute to the integral UCN production rate for a wide range of incident neutron energies [20, 21]. At low temperatures the probability for upscattering is strongly suppressed by the Boltzmann factor. Therefore, and since pure ^4He has no cross section for neutron absorption, a large density of UCN may build up in a converter with reflective walls. The storage time constant τ can of course not exceed a limit close to 900 s set by the neutron beta decay lifetime.

In a recent experiment we have demonstrated for the first time that one may efficiently extract UCN from the converter after having them accumulated therein [22]. As in past experiments [16, 17, 23, 24] the UCN production rate was found to agree reasonably well with the theoretical expectation. We have developed a new cryostat designed to keep the source portable and easy to operate. With a short cooling cycle of a few days, our system is much more flexible than an earlier apparatus [25], which was designed to be installed close to the target of a spallation source. Our present apparatus is a prototype for a future UCN source to be installed at a strong cold neutron beam of a high-flux reactor, where no extraordinary cooling power is required. A particular feature is the vertical extraction of the UCN through a cold mechanical valve situated above the helium bath. In contrast to a previous attempt to extract accumulated UCN horizontally [26], no gaps or windows are required in our method. To gain first experience with this system, the UCN converter vessel and extraction guide system were made of stainless steel. In the first run it enabled us to measure, with negligible background, the UCN production rate and to study the temperature-dependent storage properties of the converter. Here we report new results obtained with the converter vessel coated with fluorinated grease (Fomblin). It has good reflection properties for UCN and, to our very surprise, positively influences the extraction time constant. Moreover, a time-of-flight (TOF) measurement of the incident cold neutron beam helped to improve the comparison with the theoretical UCN production rate.

2 Apparatus

The apparatus was already described in some detail in ref. [22]. The converter vessel has a volume of about 2.4 liters, made from an electropolished stainless steel tube with length 696 mm and inner diameter 66 mm, closed by Ni windows on both ends. The lowest temperature attained for the completely filled vessel was $T = 0.72$ K in the previous run, and 0.82 K in the present. The maximum kinetic energy of UCN storable in the vessel is defined by the Fermi potential $V_{\text{F,wall}}$ of the wall material, which is (184 ± 4) neV for the stainless steel used (and 252 neV for the Ni windows), from which one has to subtract the Fermi potential of the superfluid helium ($V_{\text{F},^4\text{He}} = 18.5$ neV). Due to a small neutron absorbing aperture with diameter 33 mm placed at the entrance window to the converter, chosen in order to avoid activation of the vessel in these first test experiments, the volume intersected by the cold neutron beam ("UCN production volume") was $V_p = 595$ cm^3 , only. In the experiments reported here, the inner surface of the vessel was coated with a thick layer of fluorinated grease (Fomblin) with a Fermi potential of (115 ± 10) neV. UCN were extracted as previously through a flapper valve situated above the superfluid helium in the "T" section of the storage tube, connecting the (uncoated) extraction line made of electropolished stainless steel to a ^3He -gas UCN detector. The present experiments were again performed at the neutron guide "NL1" at the Munich research reactor FRM II, using the same neutron beam collimation as in the previous setup [22].

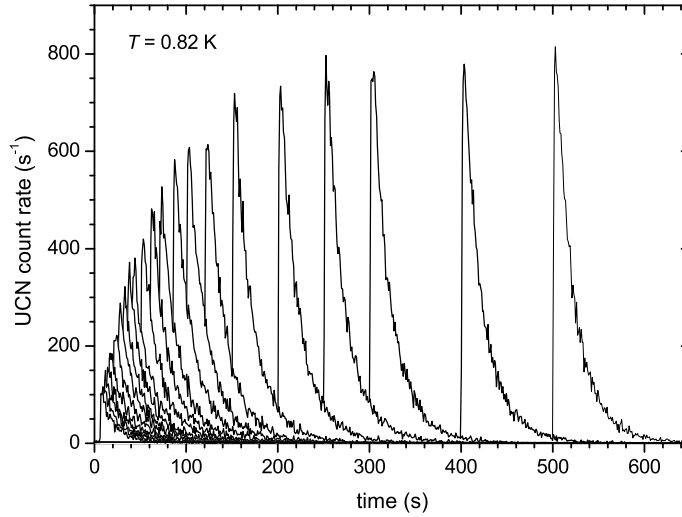


Figure 1: Time histograms of UCN count rates, measured in "buildup mode" (see text) at 0.82 K for 23 different accumulation times t_0 .

3 Definition of time constants and measurements

UCN storage and extraction from the converter can be characterised by various time constants. The storage time constant τ quantifies the temporal decrease of UCN in the closed vessel. The rate τ^{-1} contains a contribution τ_0^{-1} due to wall collisions, absorbing impurities, and UCN escaping through small holes in the vessel. This contribution does not depend on temperature T but on the kinetic energy E of the UCN. In addition there are the rates for the T -dependent UCN upscattering, and for neutron beta decay¹,

$$\tau^{-1} = \tau_0^{-1}(E) + \tau_{\text{up}}^{-1}(T) + \tau_{\beta}^{-1}. \quad (1)$$

The emptying time constant τ_e quantifies the temporal decrease of UCN in the vessel with the UCN valve open. Therefore,

$$\tau_e^{-1}(T, E) = \tau^{-1}(T, E) + \tau_A^{-1}(E), \quad (2)$$

where τ_A is the time constant for UCN passage through the extraction hole with area A . From these time constants we may derive the detection probability W for a UCN created in the converter vessel. It characterises the efficiency of the whole system including extraction and is given by

$$W = \varepsilon \frac{\tau_A^{-1}}{\tau_e^{-1}} = \varepsilon \frac{\tau - \tau_e}{\tau}, \quad (3)$$

where the factor ε describes losses in the extraction line and imperfect detector efficiency. The conversion process employed in the present experiments produces a broad spectrum of low-energy neutrons. Only neutrons with energies below $V_{\text{F,wall}} - V_{\text{F},^4\text{He}}$ are trapped inside the converter vessel, whereas those with higher energies quickly escape. The spectrum of the neutrons remaining in the vessel is shaped due to wall losses. The energy dependence of these losses is

¹In the present experiments this contribution forgotten to mention in ref. [22] was still small compared to the sum of the other rates.

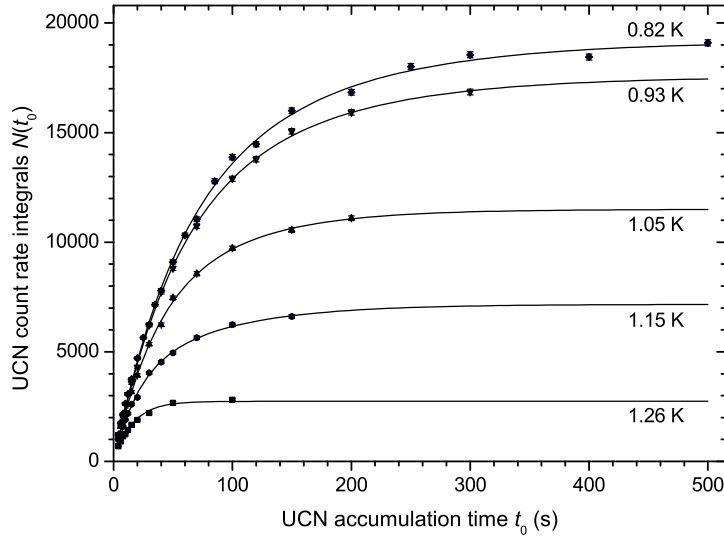


Figure 2: UCN count rate integrals as a function of the accumulation time t_0 . The fit to the data points at $T = 1.26$ K is performed with the single-exponential function defined in eq.(4). The results for the fitting parameters τ and N are employed in section 4 to determine the UCN production rate. The fits to the data points for lower temperatures employ double exponentials and serve as guides to the eye.

due to the increase with UCN energy of both the average loss probability per wall collision and the frequency of wall collisions. Thus the largest losses occur for those neutrons with energies close to the Fermi potential. However, if on a given time scale the change of the energy spectrum stays sufficiently small, the time constant τ is still well defined. This is found to be a reasonably good approximation for the accumulation of UCN at high temperatures, where the energy-independent $\tau_{\text{up}}^{-1}(T)$ dominates the rate τ^{-1} (see eq.(1)). At lower temperatures we may still deduce values for the various τ s (still calling them "time constants"), and study how they depend on the times of UCN accumulation and trapping.

The time constants τ and τ_e can be obtained from measurements in the "buildup mode". There, the closed converter is first irradiated with cold neutrons for a time t_0 , after which the beam is shut off and simultaneously the UCN valve is opened. During the whole process a time histogram of the UCN count rate is recorded. Figure 1 shows a series of such histograms for different accumulation times t_0 , which was obtained for the lowest temperature of the converter in the present runs. The decrease of the UCN count rate while emptying the vessel proceeds with a pure single-exponential; fits of the whole decay in each of the histograms with $A \exp(-t/\tau_e)$ always resulted in a reduced χ^2 close to unity and provided a value for τ_e for each of the histograms.

Figure 2 shows the integrals $N(t_0)$ of all histograms measured. It demonstrates the saturation behaviour and the strong T dependence of UCN accumulation for the range of temperatures investigated. If the time constant τ is sufficiently well defined, one can fit to the data the single-exponential buildup function

$$N(t_0) = N(1 - \exp(-t_0/\tau)). \quad (4)$$

For the highest temperature, $T = 1.26$ K, this fit has a reduced χ^2 of 3.5. The fit becomes

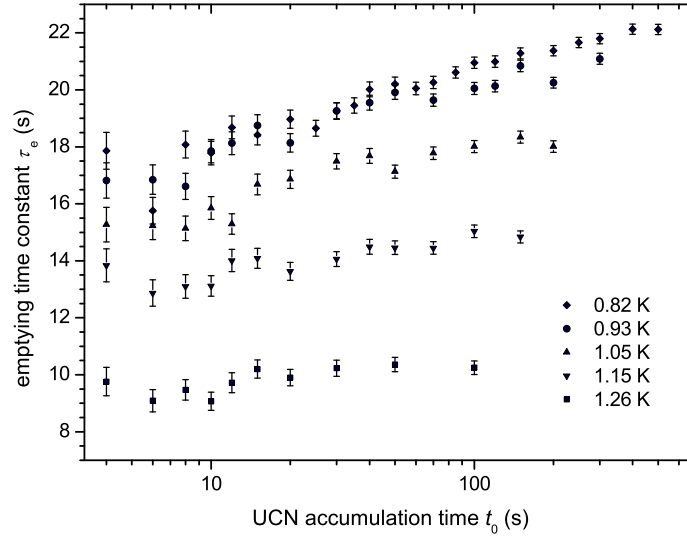


Figure 3: Emptying time constants τ_e as a function of the UCN accumulation time t_0 , determined from the histograms of all buildup-mode measurements for the various converter temperatures.

increasingly worse for the lower temperatures. As visible in fig. 3, there is also a dependence of τ_e on t_0 , which becomes increasingly pronounced for lower temperatures. These facts indicate the increasing influence of the first term in eq.(1) with decreasing temperature. The fit of a constant value to the data for τ_e at 1.26 K has $\chi^2 = 2.3$. For later use we summarise below the results of the fits to the data at this temperature, and the corresponding value for τ_A , obtained with eq.(2) and the value for τ from eq.(5). The uncertainties stated for τ , N , and τ_e have been scaled to provide a fit with $\chi^2 = 1$:

$$\begin{aligned}
 \tau &= (15.79 \pm 0.82) \text{ s}, \\
 N &= 2740 \pm 73, \\
 \tau_e &= (9.92 \pm 0.15) \text{ s}, \\
 \tau_A &= (26.7 \pm 2.6) \text{ s}.
 \end{aligned}
 \tag{5}$$

From the buildup-mode measurements as described before we cannot extract values for τ as a function of the time the UCN stay trapped in the vessel. In order to demonstrate this dependence we employed a variant of such measurements with a delayed extraction. There again the converter is first irradiated with cold neutrons while the UCN valve stays closed. After an accumulation time t_0 the beam is shut, but the UCN valve is opened only after a delay time t_d . A series of such measurements provides histograms for the same t_0 but various t_d . A plot of the count rate integrals of two series, for $t_0 = 25$ s and 250 s, is shown in fig. 4. Values for τ can be obtained from single-exponential fits to a group of several data points. The variation of slope of the curves shown in fig. 4 indicates a more pronounced increase of τ with t_d when UCN are accumulated for a longer time. This is to be expected, since after $t_0 = 250$ s the relative abundance of slower UCN with respect to the (more abundant) faster ones will be significantly higher than after 25 s, due to the longer storage time constant of the slower UCN. Their presence in the vessel then becomes better visible in the delayed-extraction experiments.

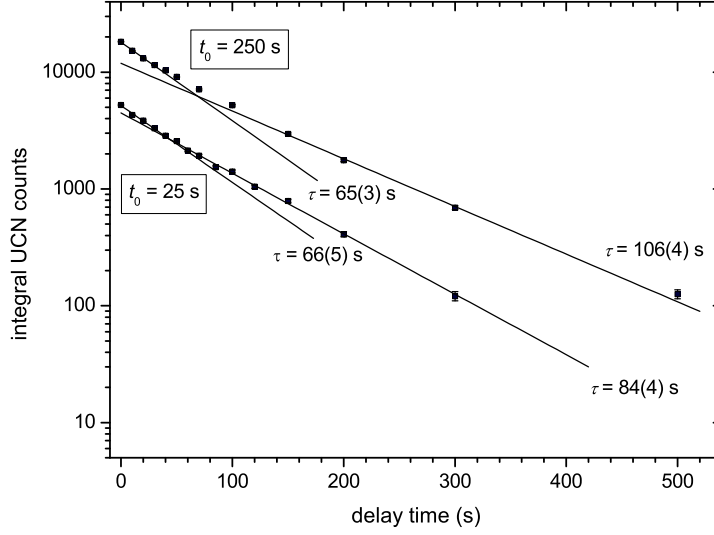


Figure 4: Count rate integrals of the measurements at 0.82 K with delayed extraction after UCN accumulation times $t_0 = 25$ s (lower data points) and 250 s (upper data points). The solid lines are fits of single exponentials to the four first, respectively, four last data points. The corresponding time constants τ derived from these fits are also shown.

T [K]	1.26	1.15	1.05	0.93	0.82
\dot{N}_c [s $^{-1}$]	176 ± 2.6	227.6 ± 2.7	257.5 ± 1.4	278.3 ± 2.3	286.6 ± 1.6
W/ε [%]	37.1 ± 3.4	48.0	54.3	58.7	60.4

Table 1: Measured T -dependent count rates obtained in continuous-mode measurements, and values deduced for W/ε (see text).

Values for τ can also be deduced from the integral UCN counts for only two different delay times, using the relation

$$\tau = \frac{t_{d2} - t_{d1}}{\ln N(t_{d1}) - \ln N(t_{d2})}. \quad (6)$$

Figure 5 shows such an analysis for the series with $t_0 = 250$ s. The strong variation of time constants from one to two minutes demonstrates indeed how much at 0.82 K the spectrum of the UCN remaining in the vessel is shaped by the energy dependence of the wall collisions during trapping.

In a third type of measurements, called "continuous mode", we measured the steady state count rate \dot{N}_c with the converter irradiated for a long time with cold neutrons while the UCN valve stayed open. The count rates measured are listed in Table 1. In the next section we will use \dot{N}_c and W/ε at 1.26 K as input to determine the UCN production rate. For the purpose of illustration we present in Table 1 also values for W/ε derived from $W(T_i) = W(1.26 \text{ K})\dot{N}_c(T_i)/\dot{N}_c(1.26 \text{ K})$, which assumes that the same UCN spectrum prevails for the different temperatures. Remember that, as obvious from fig. 3, this assumption is only valid as an approximation. The values stated for the lower temperatures might represent the true value with a relative error in the order of 10 %.

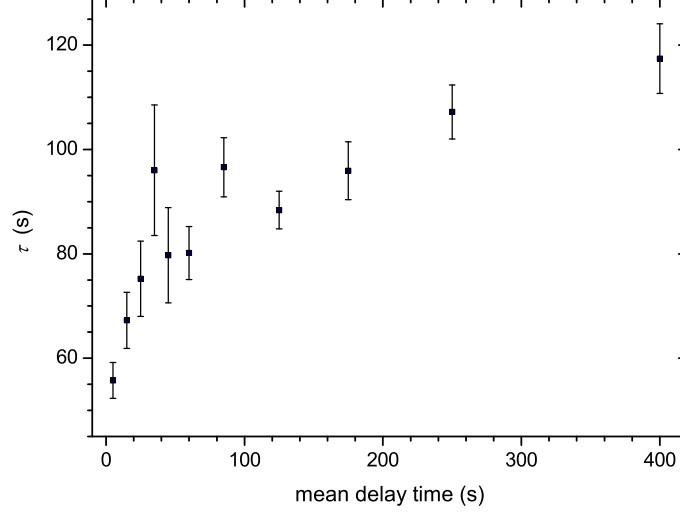


Figure 5: UCN storage time constants deduced from eq.(6), using adjacent data points from the measurements with delayed extraction shown in fig. 4. The abscissa is given by the mean of the two adjacent delay times, $(t_{d_{i+1}} + t_{d_i})/2$.

4 UCN production rate

The UCN production rate can be inferred from two methods, for which we employ the data at the highest temperature, $T = 1.26$ K, where the time constants are sufficiently well defined. First, as in the analysis of our last experiments [22], we may use the stationary "continuous mode" count rate \dot{N}_c . The corresponding UCN production rate density P_1 is given by

$$P_1 = \frac{\dot{N}_c}{V_p W}. \quad (7)$$

Using the values for \dot{N}_c and W/ε stated in Table 1 and the value $V_p = 595$ cm³ for the UCN production volume, we obtain

$$\varepsilon P_1 = (0.797 \pm 0.074) \text{ s}^{-1} \text{ cm}^{-3}. \quad (8)$$

Lacking the knowledge of ε , the numerical value in eq.(8) provides a lower limit for P_1 .

A second value for the production rate density, P_2 , may be derived from the saturated UCN number N in the buildup-mode measurement, employing eq.(4). When UCN are accumulated for a long time, $t_0 \gg \tau$, the production rate equals the UCN loss rate. Denoting with N_0 the saturated number of UCN in the vessel, of which only the fraction $N = N_0 W$ is detected, this leads us to

$$P_2 = \frac{N}{W V_p \tau}. \quad (9)$$

Using the values for τ and N from eq.(5) with W/ε from Table 1 we obtain

$$\varepsilon P_2 = (0.786 \pm 0.085) \text{ s}^{-1} \text{ cm}^{-3}. \quad (10)$$

Hence, within the 10 % accuracy of these two methods, $P_1 = P_2$. We may now compare the experimental findings with the theoretical expectation. This requires knowledge of the incident

cold neutron spectrum which we measured in a separate experiment, using a time-of-flight (TOF) analysing device described in ref. [27]. It consists of a mechanical chopper and a ^3He detector with a vertical slit. During a measurement the detector was moved horizontally in order to integrate over the whole divergence of the beam. The entrance aperture of the TOF analyser was placed at the position where previously was situated the entrance window to the converter vessel. We thus determined the neutron wavelength spectrum up to $\lambda = 2$ nm (frame overlap of the chopped bunches occurred only for $\lambda > 5$ nm, where the intensity is negligibly small). The part for $\lambda \leq 1$ nm is shown in fig. 6. After the TOF measurements the spectrum was calibrated with a gold foil activation by the integral neutron flux.

The mechanism of UCN production contains contributions from single phonon emission and multiphonon processes. The single-phonon contribution to the production rate density in a helium converter with Be wall coating is $P_I = (4.55 \pm 0.25) \times 10^{-9} \text{ d}\phi/\text{d}\lambda|_{\lambda^*} \text{ s}^{-1}\text{cm}^{-3}$, where the differential flux at $\lambda^* = 0.89$ nm is given in $\text{cm}^{-2}\text{s}^{-1}\text{nm}^{-1}$ [17]. For the present situation this value needs to be corrected for the Fermi potential of the Fomblin grease, i.e. divided by $(252 - 18.5)^{3/2} / (115 - 18.5)^{3/2} = 3.76$, with an uncertainty of 0.59 due to the poor knowledge of the Fermi potential of the Fomblin grease. From the measured TOF spectrum shown in fig. 6a we find $\text{d}\phi/\text{d}\lambda|_{\lambda^*} = 5.0 \times 10^8 \text{ cm}^{-2}\text{s}^{-1}\text{nm}^{-1}$, from which we expect a single-phonon production rate density $P_I = (0.61 \pm 0.10) \text{ s}^{-1}\text{cm}^{-3}$.

The differential multi-phonon production rate density is given by

$$\frac{\text{d}P_{\text{II}}}{\text{d}\lambda} = n_{^4\text{He}} \sigma_{^4\text{He}} E_c \frac{k_c}{3\pi} \frac{\text{d}\phi}{\text{d}\lambda} s_{\text{II}}(\lambda) \lambda. \quad (11)$$

$n_{^4\text{He}}$ is the number density and $\sigma_{^4\text{He}}$ is the cross section per helium atom. E_c and k_c are the maximum kinetic energy and wavenumber of the neutrons trapped in the vessel. $s_{\text{II}}(\lambda)$ is the scattering function $s_{\text{II}}(Q, \omega)$, evaluated on the dispersion curve of the free neutron, i.e. $\omega = \hbar Q^2/(2m_n)$, with $Q = 2\pi/\lambda$. It is modeled² to fit data from [28, 29] which is also shown in fig. 6a. Integration of $\text{d}P_{\text{II}}/\text{d}\lambda$ from 0.52 nm to ∞ for the measured incident differential flux provides an estimated lower limit of $0.42 \text{ s}^{-1}\text{cm}^{-3}$ for the integral multi-phonon production rate P_{II} . Difficult to estimate is the contribution to P_{II} from the wavelength range $\lambda < 0.52$ nm. First, there is no direct experimental information available about $s_{\text{II}}(\lambda)$ and second, this region has a strong weight due to the relatively large $\text{d}\phi/\text{d}\lambda$. The λ -dependent measurements of UCN production by Baker and colleagues [17] indicate that a multi-phonon contribution exists also from the region $0.4 \text{ nm} \leq \lambda \leq 0.52 \text{ nm}$. The measured integral multi-phonon and single-phonon production rates were found to add up evenly to the measured integral production rate [30]. This provides an experimental hint that for $\lambda < 0.4$ nm the multi-phonon contribution might be negligible. The fit function shown in fig. 6 was therefore designed to extend the range of available data with a smooth decrease to zero at 0.4 nm. Integration of $\text{d}P_{\text{II}}/\text{d}\lambda$ using the complete fitting function for $s_{\text{II}}(\lambda)$ yields $P_{\text{II}} = 0.56 \text{ s}^{-1}\text{cm}^{-3}$. Estimating the uncertainty of P_{II} as the difference between the values with and without inclusion of the contribution from the range $0.4 \text{ nm} \leq \lambda \leq 0.52 \text{ nm}$, we may expect a total UCN production rate of

$$P_{\text{th}} = (0.61 \pm 0.10)_I \text{ s}^{-1}\text{cm}^{-3} + (0.56 \pm 0.14)_{\text{II}} \text{ s}^{-1}\text{cm}^{-3} = (1.17 \pm 0.17) \text{ s}^{-1}\text{cm}^{-3}. \quad (12)$$

Evidently, more experimental information about $s_{\text{II}}(\lambda)$ is required for a secure prediction of the UCN production rate induced by a white cold neutron beam. Note that this is particularly necessary for the region of short wavelengths, whereas the contribution to P_{II} from the region $\lambda > 0.8$ nm is only $0.03 \text{ s}^{-1}\text{cm}^{-3}$. We also note that predictions from the two model calculations [20, 21] for the region $\lambda < 0.52$ nm give mutually inconsistent results.

²More details about the modeling of $s_{\text{II}}(\lambda)$ will be described in a forthcoming paper.

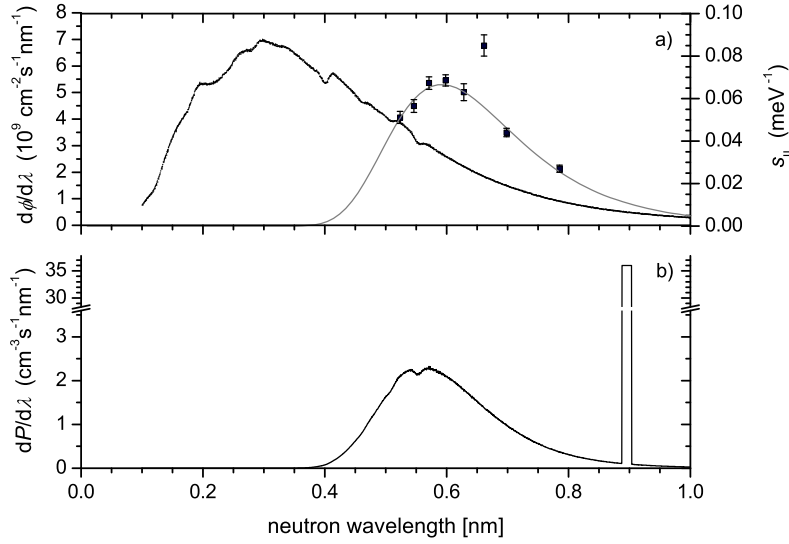


Figure 6: a) Differential flux at the neutron guide NL1 of FRMII (left scale), and measured scattering function $s_{II}(\lambda)$ and fitted curve (right scale). The data point above the curve is due to the roton-maxon resonance in the multi-phonon dynamic structure factor. b) Calculated differential production rate, including single- and multi-phonon contributions, employing the measured $d\phi/d\lambda$ and the fit function for $s_{II}(\lambda)$ from fig. 6 a).

5 Discussion and conclusions

Our experiments demonstrate the feasibility of a versatile, intense UCN source at a cold neutron beam, providing UCN for experiments at room temperature. With the present prototype, at $T = 0.82 \text{ K}$ and using a small extraction hole with area $A = 2 \text{ cm}^2$, about 60 % of the UCN produced in the Fomblin grease-coated converter vessel reached the detector. Compared to our previous experiment with the uncoated electropolished stainless steel converter vessel [22], the count rates of accumulated UCN observed immediately after opening the UCN valve, and also the integral UCN counts were almost a factor three higher, despite the lower Fermi potential of the Fomblin wall coating. This increase of UCN output is due to longer storage time constants and a faster passage of the UCN through the extraction hole. Values for τ_A as derived from eq.(2) can be compared to the gas-kinetic equation

$$\tau_A = \frac{4V}{vA}, \quad (13)$$

where we insert $V \approx 2.4 \text{ l}$ for the volume of the converter vessel, and the mean velocity v of the trapped UCN, which, for a uniform distribution in velocity space, is given by $v = 3v_{\text{max}}/4$ with $m_n v_{\text{max}}^2/2 = V_{\text{F,wall}} - V_{\text{F,}^4\text{He}}$, where m_n is the neutron mass. From the data for the Fomblin-coated vessel at $T = 1.26 \text{ K}$ we thus expect $\tau_A = 14.8 \text{ s}$. The measured value, $\tau_A = (27 \pm 3) \text{ s}$, is much closer to the expected value than in our earlier experiments with the uncoated converter vessel. There, from eq.(13) with the Fermi potential of stainless steel we may expect $\tau_A = 11.3 \text{ s}$ but measured $\tau_A = (58 \pm 13) \text{ s}$. A possible explanation is that UCN trajectories do not explore quickly enough the available phase space in the electropolished vessel, a hypothesis already raised earlier by W. Mampe and co-workers for their liquid-wall UCN bottle experiment [31]. Thus a certain roughness of the vessel walls seems to be necessary for efficient extraction of the UCN through a small hole. This should be taken into account in the design of a large converter

vessel in order to keep the time needed to extract the UCN reasonably short.

The UCN production rate densities determined with two partly independent methods agree very well, $\varepsilon P_1 = (0.80 \pm 0.08) \text{ s}^{-1}\text{cm}^{-3}$, and $\varepsilon P_2 = (0.79 \pm 0.09) \text{ s}^{-1}\text{cm}^{-3}$. In our earlier experiment with the uncoated stainless steel vessel, we obtained the values $\varepsilon P_1 = (0.91 \pm 0.21) \text{ s}^{-1}\text{cm}^{-3}$, and $\varepsilon P_2 = (1.09 \pm 0.25) \text{ s}^{-1}\text{cm}^{-3}$. The accuracy of the comparison has thus been improved by more than a factor two in the present experiments. More important, also the comparison with the theoretical expectation has been improved by virtue of a dedicated TOF analysis of the cold neutron beam spectrum, due to which we may expect the UCN production rate density $P_{\text{th}} = (1.17 \pm 0.17) \text{ s}^{-1}\text{cm}^{-3}$, with single- and multi-phonon contributions of similar size. Since the detector and extraction efficiencies, expressed by the factor ε , cannot be expected as perfect, the measured values come indeed very close to P_{th} .

In the present experiments the saturated UCN density, normalised to the production volume V_p did not exceed 46 UCN per cm^3 . To avoid activation of the stainless steel vessel only a quarter of the total volume was irradiated with the cold beam. Also for practical reasons, significant divergence losses due to beam collimation were accepted. In addition, for simplicity of these first tests, we did not yet employ a wall material with low neutron absorption and a high Fermi potential of 250 neV or beyond, such as Be, BeO or diamond-like carbon [32, 33]. There one will gain in UCN density both due to the larger energy of the storable neutrons and due to a larger storage time. In order to produce a larger total number of UCN, the converter vessel can be made much longer, due to the low cross section of $27 \times 10^{-27} \text{ cm}^2$ for neutrons with wavelength 0.89 nm [34], corresponding to a mean free path of about 17 m. With a properly designed ^4He converter and using an existing intense cold neutron beam at a high flux reactor, UCN densities in the order 10^4 per cm^3 with a total UCN number up to several 10^8 seem within reach.

Acknowledgements: We are very grateful to the head of the Physics Department E18, Prof. S. Paul, and to the scientific director of the FRM II, Prof. W. Petry, for supporting this development. We also thank I. Altarev, A. Müller and W. Schott for some useful discussions. We gratefully acknowledge the help of M. Pfaller and his team from the central mechanical workshop of the physics faculty for careful manufacturing of many cryostat components. This work has been funded by the German BMBF (contract number 06MT250).

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